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Project Title: Excitation and Ionization of Ions by Electron Impact

Project No: E-21-688

Project Director: Dr. Robert K. Feeney

Sponsor: Energy Research and Development Administration
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E-21-688

REPORT NO. ORO-3027-38

Technical Progress Report

Project No. E-21-688

Covering the Period
September 1, 1976 to May 31, 1977

*THE EXCITATION AND IONIZATION OF IONS
BY ELECTRON IMPACT*

By R.K. Feeney
D.W. Baggett
D.W. Hughes
G.W. Rivers
W.E. Sayle

Contract No. EY-76-S-05-3027

U. S. Energy Research and Development Administration
Oak Ridge, Tennessee

31 May

1977



School of Electrical Engineering
GEORGIA INSTITUTE OF TECHNOLOGY
Atlanta, Georgia

REPORT NO. ORO-3027-38

PROJECT NO. E-21-688

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U. S. Energy Research and Development Administration

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ABSTRACT

This effort is devoted to experimental measurements of electron impact excitation and ionization cross sections of ions. The cross sections of interest are those of importance in the diagnostics of CTR plasmas. Current tasks include: (a) the completion of absolute measurements of the electron impact double ionization cross sections for Na^+ , K^+ , Rb^+ , Cs^+ , and Tl^+ ions; (b) the development of a laboratory-size ion source of multiply-charged ions to be used in the measurement of electron impact excitation and ionization cross sections; and (c) the completion of absolute measurements of the electron impact excitation of Li^+ ions.

Preliminary measurements of the electron impact double ionization cross sections of Na^+ , K^+ , Rb^+ , Cs^+ , and Tl^+ ions have been completed. Measurements were made over the range of electron energies from the respective threshold values to approximately 1000 eV. Peak cross sections were found to vary from $7.2 \times 10^{-19} \text{ cm}^2$ for Na^+ to $3.5 \times 10^{-17} \text{ cm}^2$ for Cs^+ . The data were obtained with a crossed beam apparatus operating with modulated beams.

A PIG-type source of multiply charged ions is undergoing final development. The source is of laboratory size and is compatible with existing collision apparatus. The previous problem with inadequate magnetic field has been solved. Spectroscopic techniques verified the production of ions of charge state C^{4+} when CO_2 was used as the source gas. Some difficulty has been encountered in extracting

adequate ion currents. Also under investigation is the optimum technique for the production of metal ions with the PIG-type ion source.

The electron impact excitation cross sections for the 2^1S-2^3P transition in Li^+ ion is being measured by observation of the 2^3P-2^3S radiation. These measurements were to facilitate the optimization of the experimental apparatus while allowing a parallel development of a source of multiply charged ions. A new collision apparatus was to be used for these measurements, but at the direction of ERDA, construction of the apparatus was postponed in order to mount a larger ion source development effort. Consequently, the current Li^+ measurements were made in the older apparatus which suffered from several shortcomings. Preliminary data have been taken but are not reported herein because of large experimental scatter.

SECTION I

DISCUSSION OF PROGRESS

This summary covers progress made during the current period of September 1, 1976 to May 31, 1977. The general goal of this work is the measurement of electron impact ionization and excitation cross sections that are of importance in near- and far-term plasma diagnostics related to the CRT program. The specific programs involved in the research effort as extracted from the currently active proposal are: (a) completion of the measurements of the absolute cross sections for double ionization of Cs^+ , Tl^+ , K^+ , and Na^+ ions by electron impact over the range of incident electron energies from below threshold to approximately 2000 eV; (b) the completion of the absolute measurements of the cross sections for electron impact excitation of $1^1\text{S}-2^3\text{P}$ transition by observation of the $2^3\text{P}-2^3\text{S}$ triplet in the He-like Li^+ ions; (c) measurements of the absolute cross sections for input excitation by electrons of the $1^1\text{S}-2^3\text{P}$ transition in the He-like B^{3+} and C^{2+} ions; and (d) the refinement and extrapolation of the techniques used in the above work so as to facilitate the determination of electron impact excitation and ionization cross sections for additional members of the He- and Be-isoelectronic sequences. Upon consultation with ERDA, after the above proposal was submitted, the research program was slightly modified. Because of possible conflict with other efforts in progress at ORNL, it was decided to make the following changes:

1. Change emphasis from atmospheric ions to other species which did not conflict with efforts at ORNL. The Li^+ excitation effort was to be continued and the new species were to be incorporated into the next proposal.
2. Prepare for this change by doing more extensive development of the PIG-type multiply-charged ion source with the goal of producing other types of ions.
3. Postpone the construction of a new collision apparatus and devote that effort to other proposed tasks.

The above changes were incorporated into the current research program, and new milestones generated. Progress toward the revised goals is being made at what is considered to be a satisfactory rate.

The following section briefly summarizes progress made and problems encountered in each of the following experimental areas: (a) ionization of ions; (b) excitation of Li^+ ions and apparatus development; and (c) development of a source of multiply-charged ions. Details of experimental results and discussions of apparatus are presented in appropriate appendices.

Ionization of Ions

Preliminary measurements of the electron double ionization cross sections of Na^+ , K^+ , Rb^+ , Cs^+ , and Tl^+ ions have been completed over the range of electron energies from threshold to approximately

1000 eV. This work was undertaken primarily to provide data for use in the calibration of ion beam probes.¹⁻⁵ Advanced ion beam probes that measure the spatial variation of electron temperature do so by virtue of the electron energy dependence of two different ionization cross sections. Two ions are injected simultaneously into the plasma and the doubly charged reaction products collected. As long as the electron energy is low enough so that one of the cross sections is changing significantly with energy, the ratio of the two collected product ions is a measure of the electron temperature. Another way to obtain the same information involves the use of one ion but both the single and double ionization processes.

The validity of the results obtained with these beam probes is critically dependent upon the accuracy of the appropriate cross sections. Until the present work, only the Li^+ double ionization cross sections were known.⁶ Accordingly, the present effort was undertaken to provide the needed double ionization cross sections. Most of the double ionization results have been disseminated in the form of conference presentations.

The double ionization cross sections were measured with a modified version of the crossed beam apparatus employed in our recent work.⁷ Details of the apparatus and a summary of the present experimental results are given in Appendix I.

The Excitation of Ions and Apparatus Development

This effort is devoted to the design, development and construction of apparatus and perfection of techniques leading to ultimate measurements

of the electron impact excitation cross sections of multiply charged ions. Spectroscopic methods can provide information as to the electron temperature, electron density, and ion density as well as impurity densities and losses.⁸⁻¹³ Almost all such techniques require knowledge of one or more collision cross sections.

The electron impact excitation of the 2^1S-2^3P transition in the He-like Li^+ ion by observation of the 2^3P-2^3S radiation is being measured. These measurements are being made with a modified version of the apparatus used in our previous work.^{14,15} Because these cross sections are very small and the emitted light is in a wavelength range susceptible to background signals, the initial measurement attempt was unsuccessful. It was planned to defer these measurements until an improved crossed beam apparatus was constructed. However, as outlined above, ERDA suggested that a larger effort be expended on the development of the multiply-charged ion source and therefore construction of the new apparatus was delayed. After some redesign, the signal-to-noise ratio obtainable with the older apparatus was increased to about 1.5%. This has enabled preliminary data to be obtained. The experimental scatter is still excessive, but it probably can be reduced by improved shielding techniques. Details of the Li^+ experiment including a description of the modifications applied to the old apparatus are given in Appendix II. A description of the proposed new apparatus was given in a previous Progress Report.¹⁶

Ion Source Development

While the measurements of the electron impact excitation of the Li^+ ions is intrinsically valuable and provides a useful tool for the

design and construction of a versatile experimental apparatus, the primary objective of the present work is directed to the multiply charged ions.

The multiply charged ion apparatus is composed of three functional sections; the ion source, the m/e analyzer, and the collision chamber. The design for the new collision chamber was described in a previous Progress Report.¹⁶ The following material discusses problems and progress of the multiply charged ion production and analysis system.

The ion source is of the Penning Ion Gauge or PIG type. This type of ion source has been used in nuclear accelerators to provide multiply charged heavy ions. The present source has been designed explicitly for atomic collisions research and is considerably smaller and simpler than the usual source for nuclear applications. The details of the source design were given in previous reports.

The PIG-type ion source is undergoing final development. The installation of a large 6-inch VARIAN magnet obtained from the School of Electrical Engineering solved the previous problem of insufficient magnetic field. A new power supply has allowed operation of the PIG source at arc currents of up to 5A. Spectroscopic techniques were used to determine the charge-state distribution within the ion source. These measurements verified the production of C^{4+} ions when CO_2 was used as the source gas. The extracted ion beam current has been smaller than it should be as indicated by comparison of its ion yield with those of similar sources in the literature. Efforts to realize the optimum current are in progress as are experiments to determine the best procedure for producing metal ions with the PIG-type source.

Since the operating parameters of this ion source are very close to the ORNL PIG-type source, it is believed that similar performance can be expected.

After exciting the ion source, the ion beam enters the magnetic sector m/e analyzer for ion selection. The previous problems with this analyzer have been solved. The magnetic field of 6 kG is considered adequate for all proposed work.

Details of the ion source and associated systems are presented in Appendix III.

SECTION II
PUBLICATIONS

The publication listed below were prepared or published during the current reporting period based upon completed research. Conference presentations are not listed.

1. Title: "Aluminosilicate Sources of Positive Ions for Use in Collision Experiments"
Authors: R. K. Feeney, William E. Sayle, II, and J. W. Hooper
Status: Published in Review of Scientific Instruments 47, 964 (1976).
Support: ERDA 50%, Georgia Tech 50%
2. Title: "Absolute Experimental Cross Sections for the Electron Impact Ionization of Rb^+ and Cs^+ Ions"
Authors: R. K. Feeney, William E. Sayle, II, and T. F. Divine
Status: Submitted for publication in Physical Review.
Support: ERDA 75%, Georgia Tech 25%.

APPENDIX I

IONIZATION OF IONS BY ELECTRON IMPACT

The electron impact double ionization cross sections for Na^+ , K^+ , Rb^+ , Cs^+ , and Tl^+ ions have been measured as a function of incident electron energy from below threshold to approximately 1000 eV. These results, together with a discussion of the experimental apparatus and technique are presented in this appendix. Some comparisons with available theoretical calculations are also given.

The experimental method involves the use of a crossed beam apparatus in which approximately monoenergetic beams of ions and electrons are caused to intersect at right angles in a well-defined collision region. The crossed beam technique has now become a well-established tool for the study of charged-particle, charged-particle collision processes. Several reviews that discuss the advantages and difficulties inherent in crossed beam experiments have been written and most of the early work utilizing this method critically evaluated.¹⁷⁻²¹ Accordingly, with the availability of such a complete body of reference material, a general discussion of the experimental technique will not be given here; however, those unique problems associated with the present experiment are discussed in appropriate sections.

A schematic diagram of the experimental apparatus is given in Figure A-1 and a plan view photograph is shown in Figure A-2. Singly charged ions are produced by a thermionic-type ion source and pass

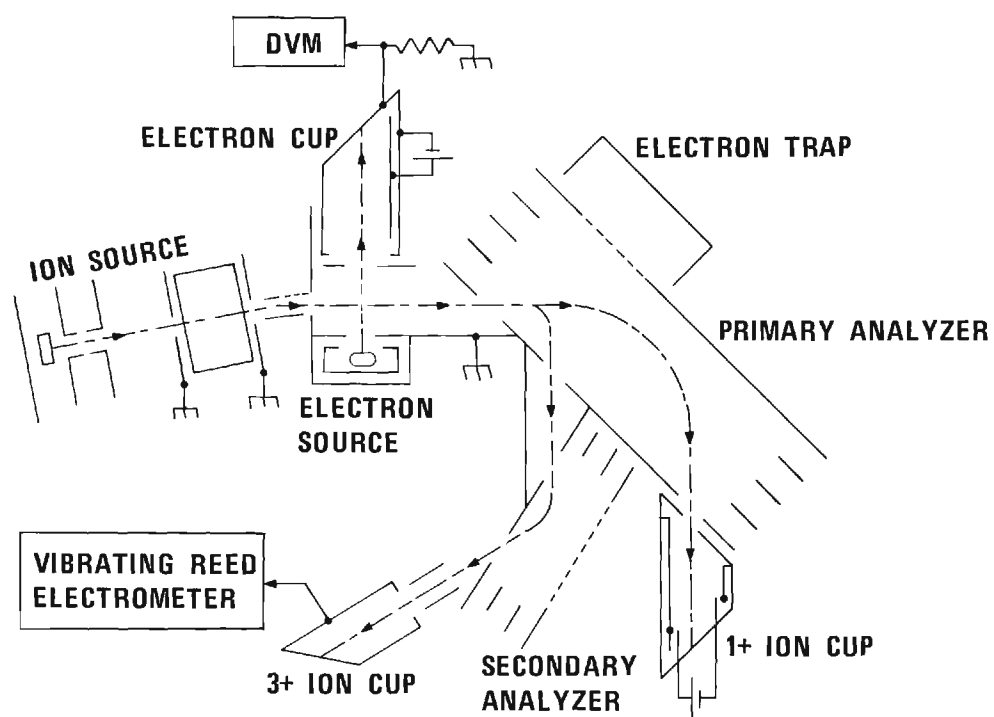


Figure A-1. Schematic Diagram of the Ionization Apparatus.

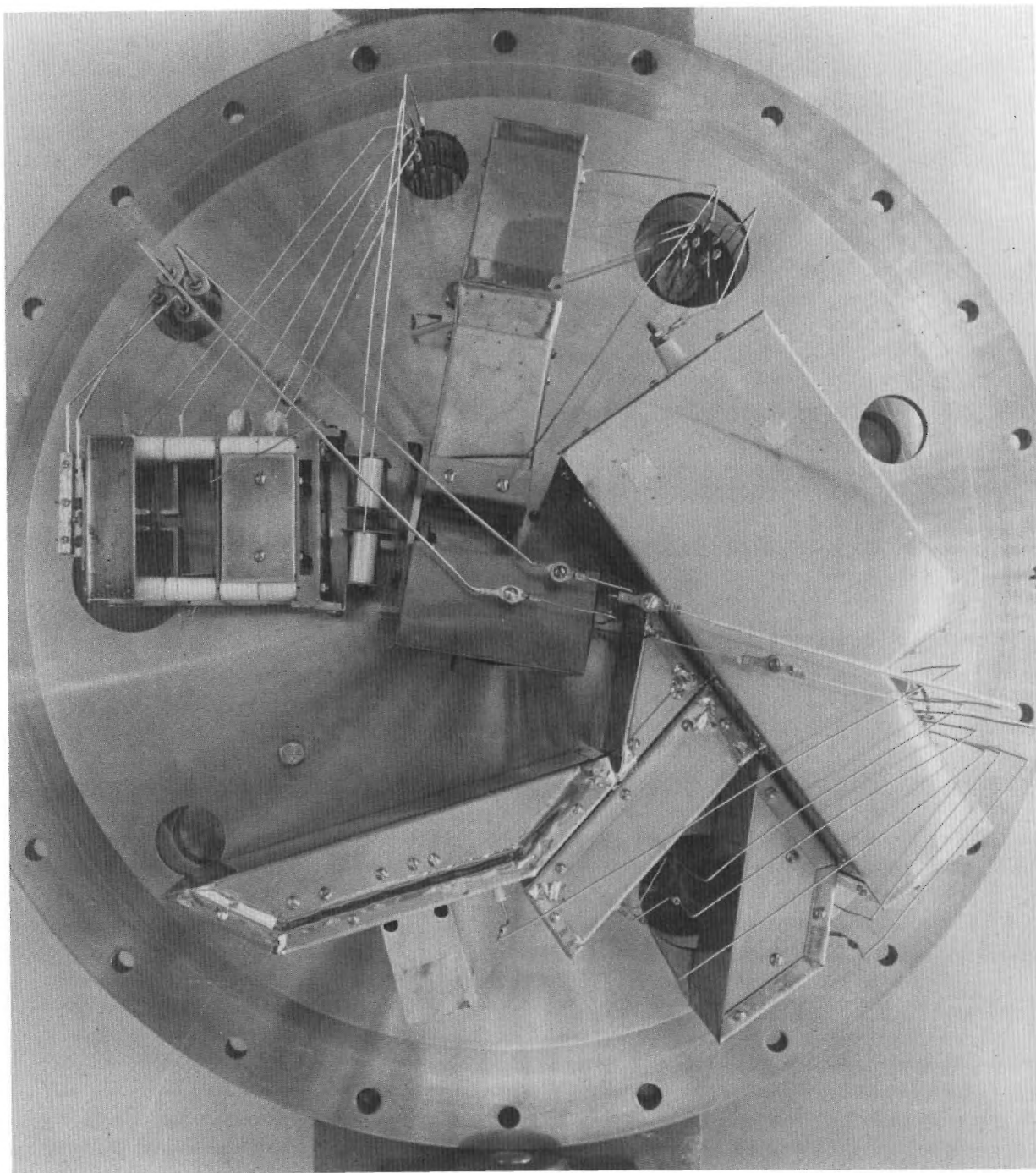


Figure A-2. Plan View Photograph of the Ionization Apparatus.

through several focusing, collimating and deflecting structures before entering the interaction region. A rectangular electron beam intersects the ion beam in the interaction region. Just prior to entering the interaction region, the two beams can be made to pass through a scanner which determines their spatial profiles. This scanner is driven from outside the vacuum chamber by a stepping motor sequenced by a preset counter. After undergoing collisions with the electrons in the interaction region, the ion beam, which now contains several charge states, passes into the large parallel plate primary electrostatic analyzer. Here the singly- and triply-charged beam components undergo initial separation.

The singly charged ion beam is diverted into a Faraday cup while the triply-charged ion beam passes into the secondary electrostatic analyzer. This analyzer removes slow ions produced by the singly charged ion beam ionizing or charge exchanging with the background gas. Such slow ions are repelled from the primary analyzer, but do not have the correct energy to traverse the secondary analyzer. Both the primary and secondary analyzers employ mesh-type back plates to reduce the chance that reflected stray electrons will be collected. Guard rings are incorporated in both for improved uniformity of the electric fields.

Upon exiting the secondary analyzer, the triply charged ion beam is collected by a Faraday cup. This cup has a secondary electron suppressor placed in front of it. In addition, permanent magnets provide a magnetic field perpendicular to the axis of the cup. This field serves as additional secondary electron suppression and also

helps to keep out scattered background electrons.

Not shown in either the diagram or the photograph, is an auxiliary ion cup which can be lowered into the path of the ion beam prior to its entering the interaction region. This cup facilitates comparison of the ion beam entering the primary electrostatic analyzer with those beams emerging from the two analyzers. The auxiliary cup has been very useful in pinpointing unusual focussing conditions that resulted in some loss from the ion beam prior to its entering the primary analyzer.

In this experiment, as in certain of our previous work,^{17,18,22} a 6L6GC beam tetrode was used as an electron source. The tube envelope was first removed except for the base portion which supported the tube structure. One side of the plate was then opened to expose the grids and beam forming structure. A clamping fixture held the tube in position allowing the electron beam to exit from the beam forming structure, through several apertures, and into the interaction region. The source is capable of providing a 2.5 mm thick electron beam of 1 mA at 150 eV and more than 10 mA at 1000 eV. The mean energy of the electron beam is about 2 eV below that set by the acceleration voltage while the energy spread is about ± 1 eV at half-maximum.

A thermionic-type ion source produced a chemically pure beam of ground state alkali ions. No differential pumping or water cooling was required with this type of source. The ion source produced a collimated (0.8 x 6.4 mm) beam of $1-3 \times 10^{-7}$ A for an operating period of several weeks. Impurity levels were always less than 0.5% of the desired ion emission current. Additional details of this and

similar ion sources are available elsewhere.²³

The electron current was determined from the voltage drop across a precision resistance as monitored with a digital voltmeter. The error in the electron current determination was less than $\pm 1\%$. The singly-charged-ion beam current was measured with a conventional electrometer while the triply-charged-ion beam current was measured with a vibrating reed electrometer operating in the rate-of-charge mode. An integrating digital voltmeter connected to the vibrating reed electrometer supplemented a chart recorder in data acquisition. The estimated error in the singly- and triply-charged ion beam currents was less than $\pm 2\%$ and $\pm 3\%$, respectively.

The experiment was operated in the pulsed beam mode.²⁴ This technique eliminated background gas density modulation effects which otherwise might occur for vacuum system pressures of greater than about 10^{-8} Torr. Both the ion beam and the electron beam were pulsed, the ion beam with a 35% duty factor and the ion beam with a 50% duty factor. By a simple change of the relative phases of the two beams, the ions and electrons could be made to cross the interaction region either in time-coincidence or in time-anticoincidence. The difference between the coincidence and anti-coincidence signals represented the electron impact ionization current.

The absolute cross sections obtained with the above apparatus are given in Figures A-3 through A-7. The data for each ion were obtained from a single operating cycle of the experiment. Our usual procedure has been to consider data preliminary until confirmed by at least two independent "runs" of the apparatus. The error bars include

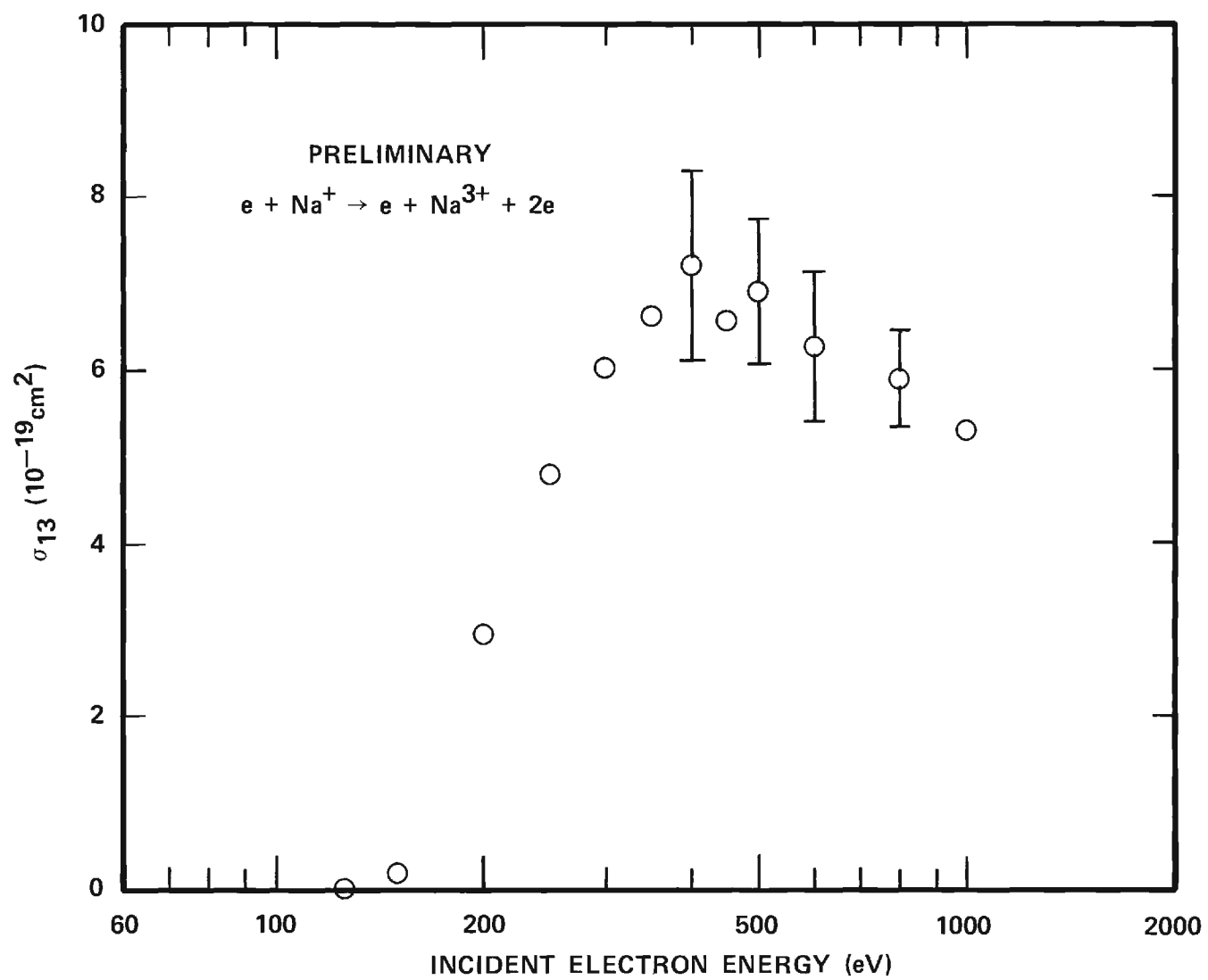


Figure A-3. Absolute Experimental Cross Sections for the Double Ionization of Na^+ Ions.

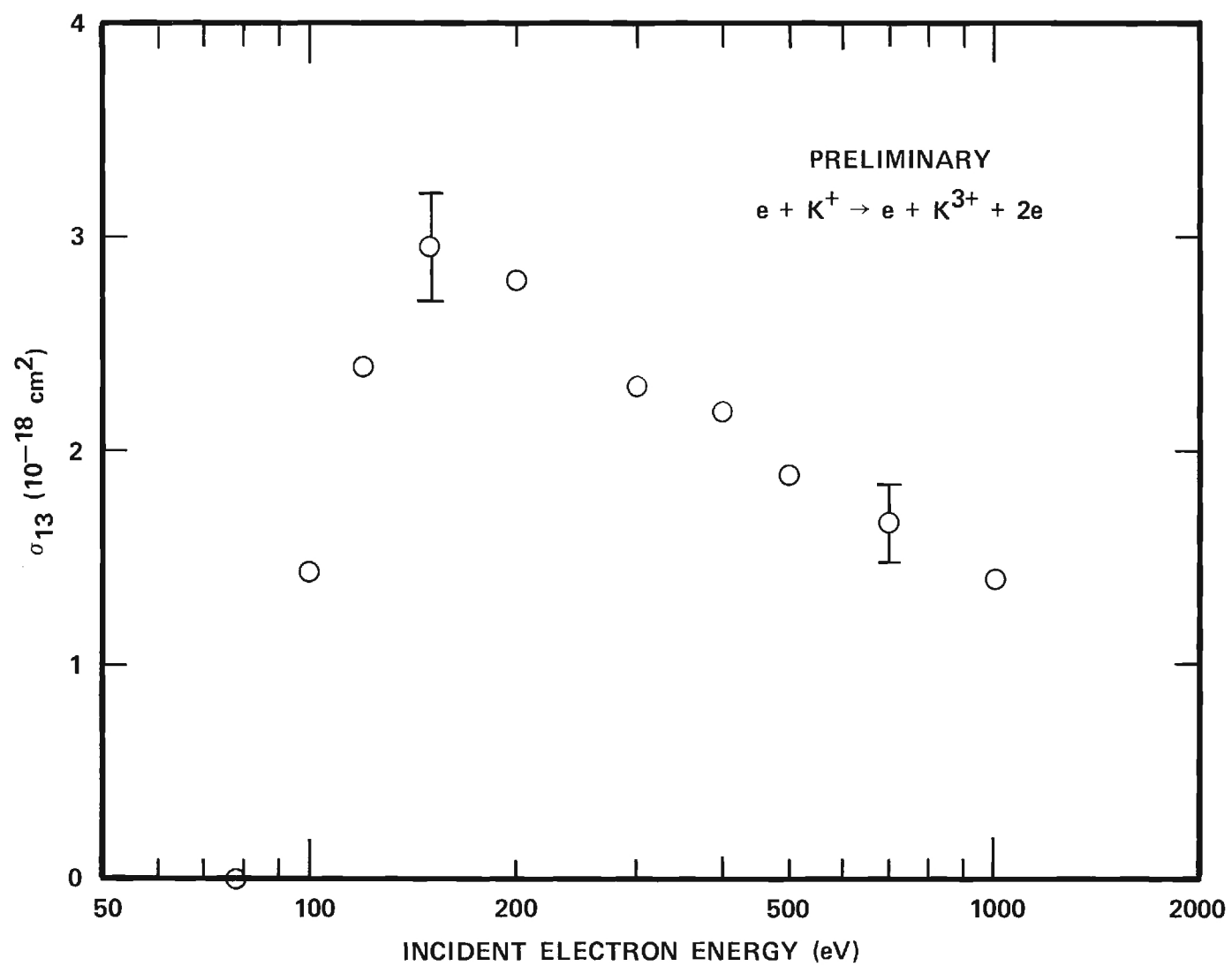


Figure A-4. Absolute Experimental Cross Sections for the Double Ionization of K^+ Ions.

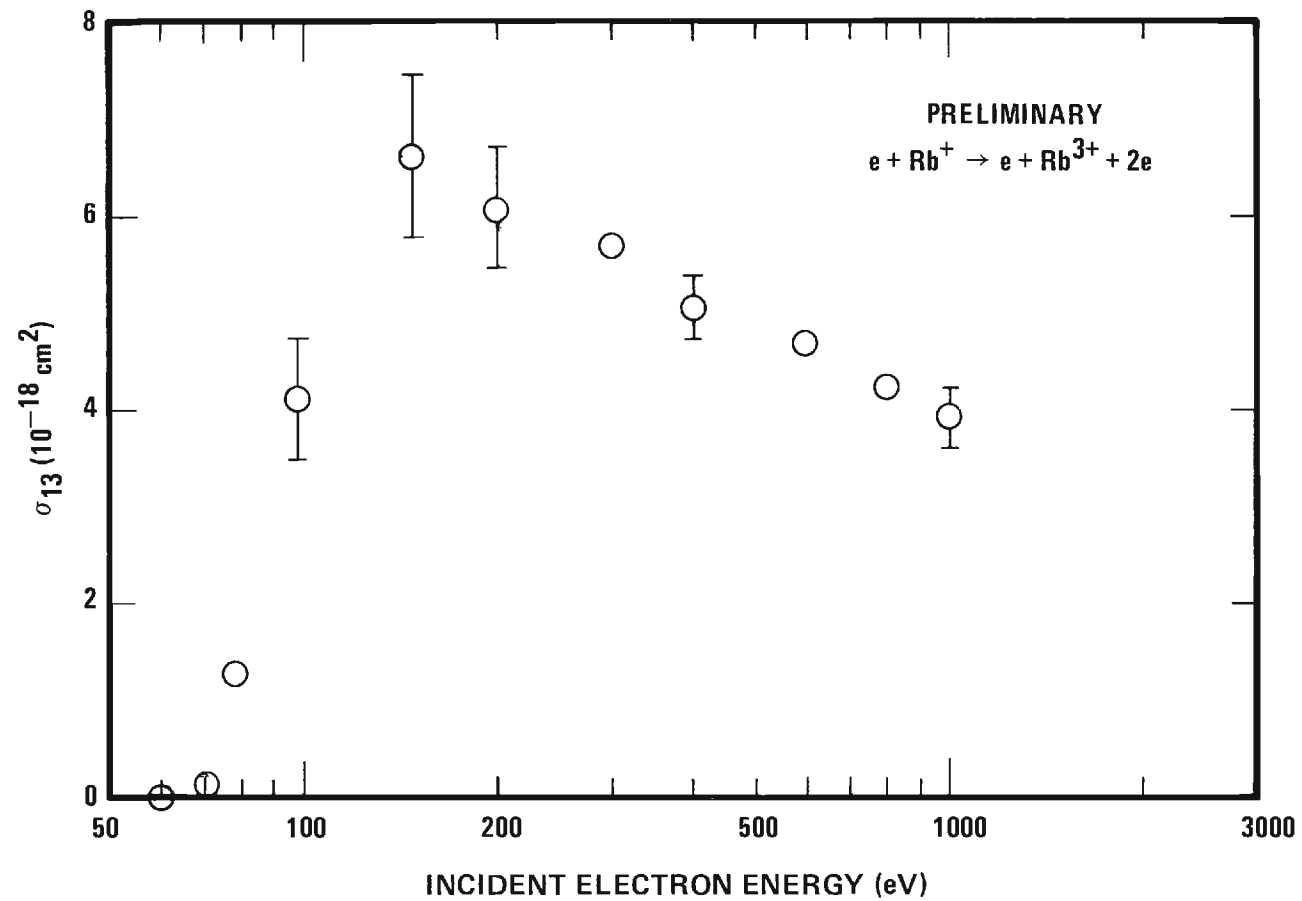


Figure A-5. Absolute Experimental Cross Sections for the Double Ionization of Rb^+ Ions.

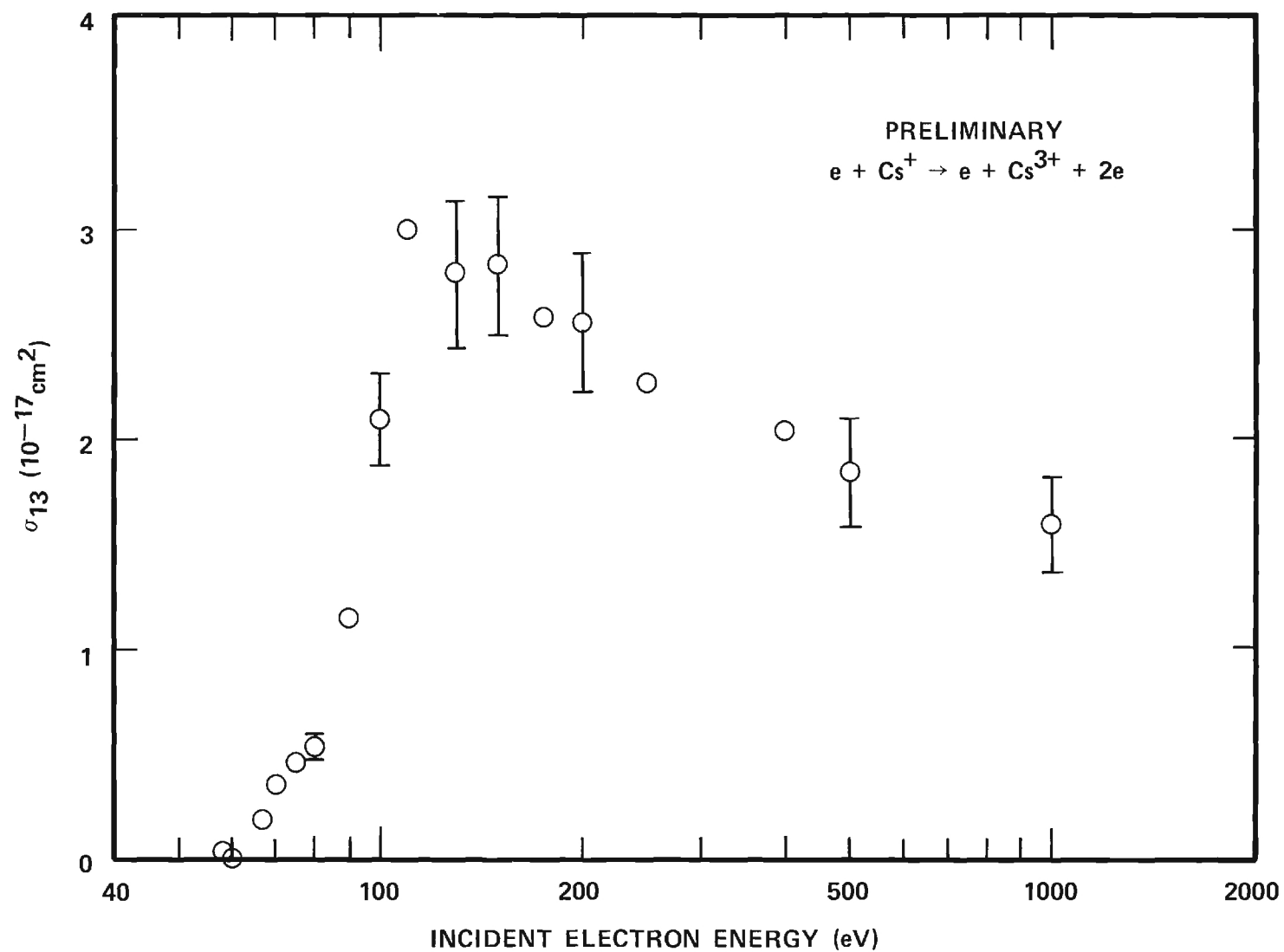


Figure A-6. Absolute Experimental Cross Sections for the Double Ionization of Cs^+ Ions.

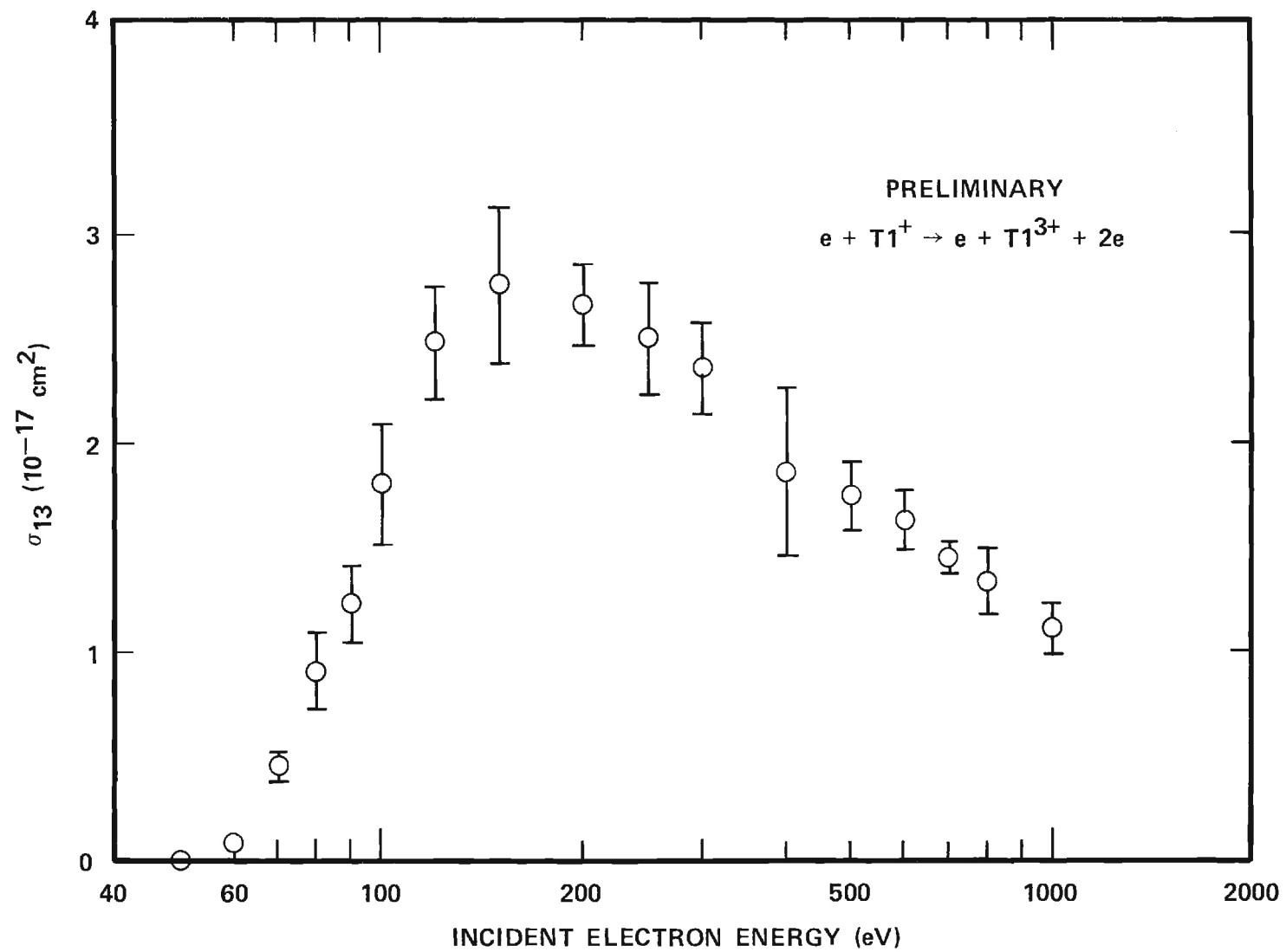


Figure A-7. Absolute Experimental Cross Sections for the Double Ionization of Tl^+ Ions.

the sum of the random error and systematic errors. Random errors were estimated from the 90% confidence limits of the mean and the systematic error is taken to be $\pm 4\%$, the uncertainty in the instrumentation calibration.

Data for the ionization of Na^+ and K^+ ions are presented in Figures A-3 and A-4, respectively. These cross sections were calculated by Tripathi and Rai²⁵ using the method of Gryzinski.²⁶ Their calculations are compared with the present results in Table A-I.

Table A-I

Ion	Peak Cross Section (cm^2)	
	measured	calculated
Na^+	7.2×10^{-19}	1.8×10^{-17}
K^+	2.9×10^{-18}	3.5×10^{-18}

The calculations would be in more systematic agreement if the exponents were reversed; however, the authors give the same results two places in their paper.

As part of the present research, we also calculated these cross sections using Gryzinski's method.²⁶ The peak cross section results for Na^+ and K^+ were, respectively, 1.7×10^{-18} and $4.9 \times 10^{-18} \text{ cm}^2$.

The present experimental results for Rb^+ , Cs^+ , and Tl^+ ions are compared with calculations using the method of Gryzinski²⁶ in Table A-II.

Table A-II

Ion	Peak Cross Section (cm ²)	
	measured	calculated
Rb ⁺	6.6×10^{-18}	6.8×10^{-18}
Cs ⁺	3.5×10^{-17}	8.0×10^{-18}
Tl ⁺	2.7×10^{-17}	4.7×10^{-17}

Since the Gryzinski technique is only a classical approximation and does not apply directly to ions, the closeness of agreement in Table A-II is of no fundamental importance. However, since plasma workers often use simple methods to obtain cross section estimates, Table A-II is valuable in indicating the order of agreement obtainable.

APPENDIX II

EXCITATION OF Li^+ IONS AND APPARATUS DEVELOPMENT

During the period covered by this report, efforts were directed to the measurement of Li^+ electron impact excitation cross sections and the development of an advanced atomic collisions measurement facility. The advanced collision facility is required for the program of multiply-charged ion excitation and ionization cross section measurements. The Li^+ experiment was undertaken to allow parallel development of the multiply charged ion source and the collision chamber. Since Li^+ ions can be produced thermionically, the collision chamber could be designed independently of the multiply charged ion facility.

As indicated previously, guidance from ERDA required that the major emphasis of the present program be directed toward the perfection of the multiply-charged ion source and the completion of the Li^+ excitation measurements. Accordingly, only a small fraction of the current year's effort was applied to the construction of the new collision chamber. Procurement of major component items was continued as was construction of the vacuum system but little detailed internal design was completed.

This appendix discusses the details of the Li^+ excitation measurements. The design of the advanced collision experiment was discussed in a previous Progress Report.¹⁶

Since the new collision apparatus was not to be completed this year, it was necessary to continue the Li^+ excitation measurements

using the older apparatus. This approach suffered from three major problems; poor electron source activation, a very large ambient light level, and background gas modulation effects.

A titanium sublimation pump was installed which resulted in a great improvement in the electron source activation. This device also reduced the vacuum chamber pressure into the 10^{-10} Torr range where the effects of background gas modulation were negligible.

A significant difficulty with the measurement of the 2^3P-2^3S transition is that the 5486 \AA visible radiation is in the same wavelength range as stray light from the ion and electron sources. The reduction in the ion source background was achieved by passing the ion beam through a 90° electrostatic analyzer prior to its colliding with the electron beam. The analyzer eliminated a direct line-of-sight path from the ion source to the interaction region. The interior of the analyzer was darkened with gold-black to reduce reflections. The position of the analyzer can be seen in the schematic drawing of the apparatus, given in Figure A-8. Since the electron source must be close to the interaction region, a 90° analyzer could not be used to eliminate the direct optical path. However, a light trap was installed directly over the photon collection lens. This reduced the scattered light from the electron source to an acceptable level. The two modifications have increased the signal-to-noise ratio to approximately 1.5% which is a great improvement over the previous figure.

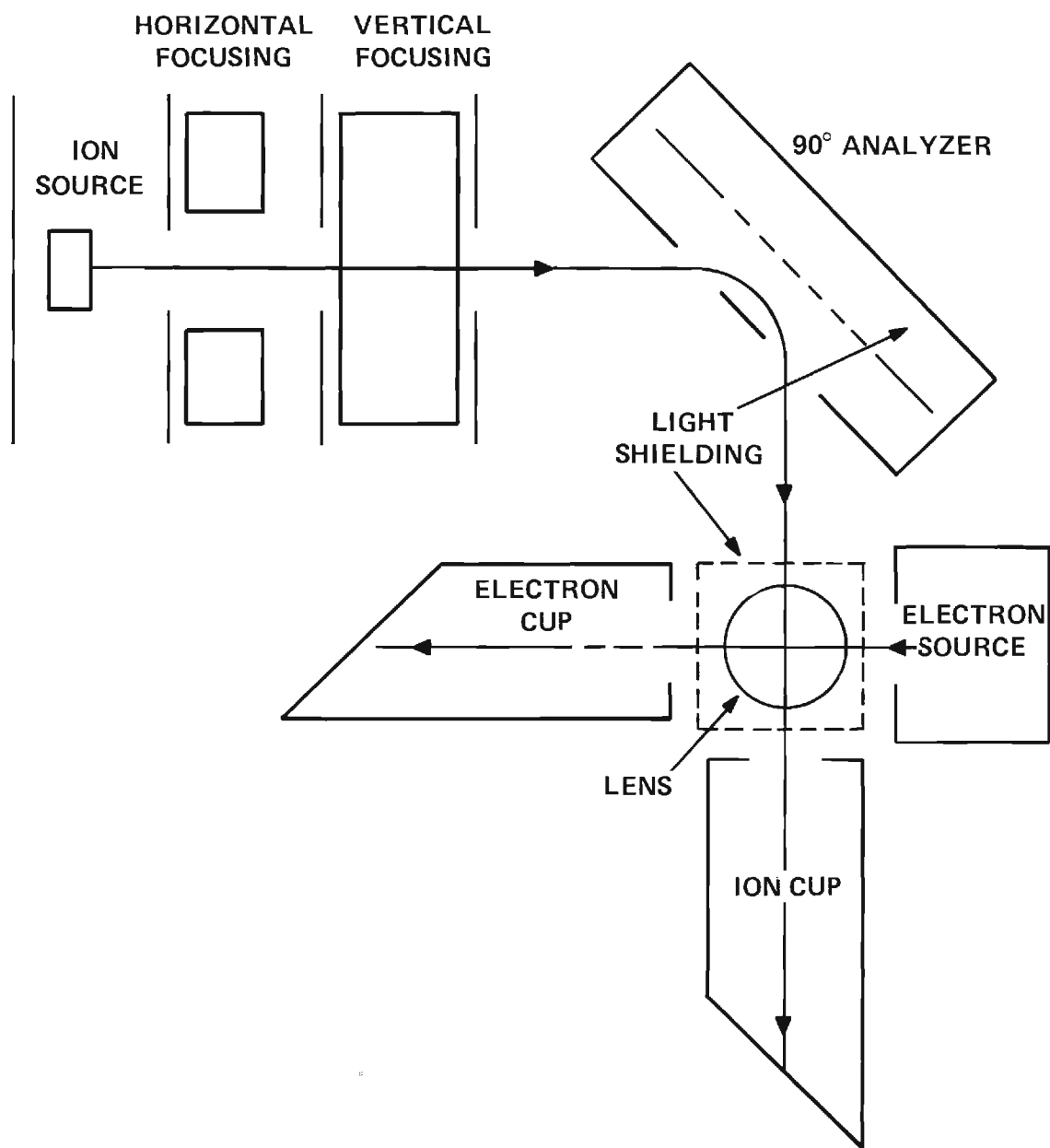


Figure A-8. Schematic Diagram of the Apparatus Used in the Present L^+ Excitation Measurements.

APPENDIX III

DEVELOPMENT OF A MULTIPLY-CHARGED ION SOURCE

A major portion of the present effort is devoted to the development of a laboratory-size ion source of multiply-charged ions. Such a source will then be used with an experimental apparatus such as described in a previous Progress Report¹⁶ to measure excitation and ionization cross sections of multiply-charged ions. This appendix discusses the multiply-charged ion source and its associated beam handling system.

Efforts during previous contract periods concentrated on an evaluation of various sources as candidates for producing ions suitable for charged-particle--charged-particle crossed beam experiments. Ion source types considered included the washer-type pulsed plasma source, the trapped-ion source, various configurations of an electron-cyclotron source, the hot-electron plasma source, the duoplasmatron source, and the Penning Ion Gauge (PIG) source. Most of the plasma-type sources were very complicated and required extensive support facilities. The duoplasmatron was rejected because of its low yield of highly-charged ions. It was concluded that the PIG source presented fewest technological risks, and could be made a suitable size for use in the laboratory.

A schematic of the multiply-charged ion source and the associated m/e analyzer is shown in Figure A-9 and a photograph is given in Figure A-10. The PIG source is mounted in one port of the cross-shaped vacuum

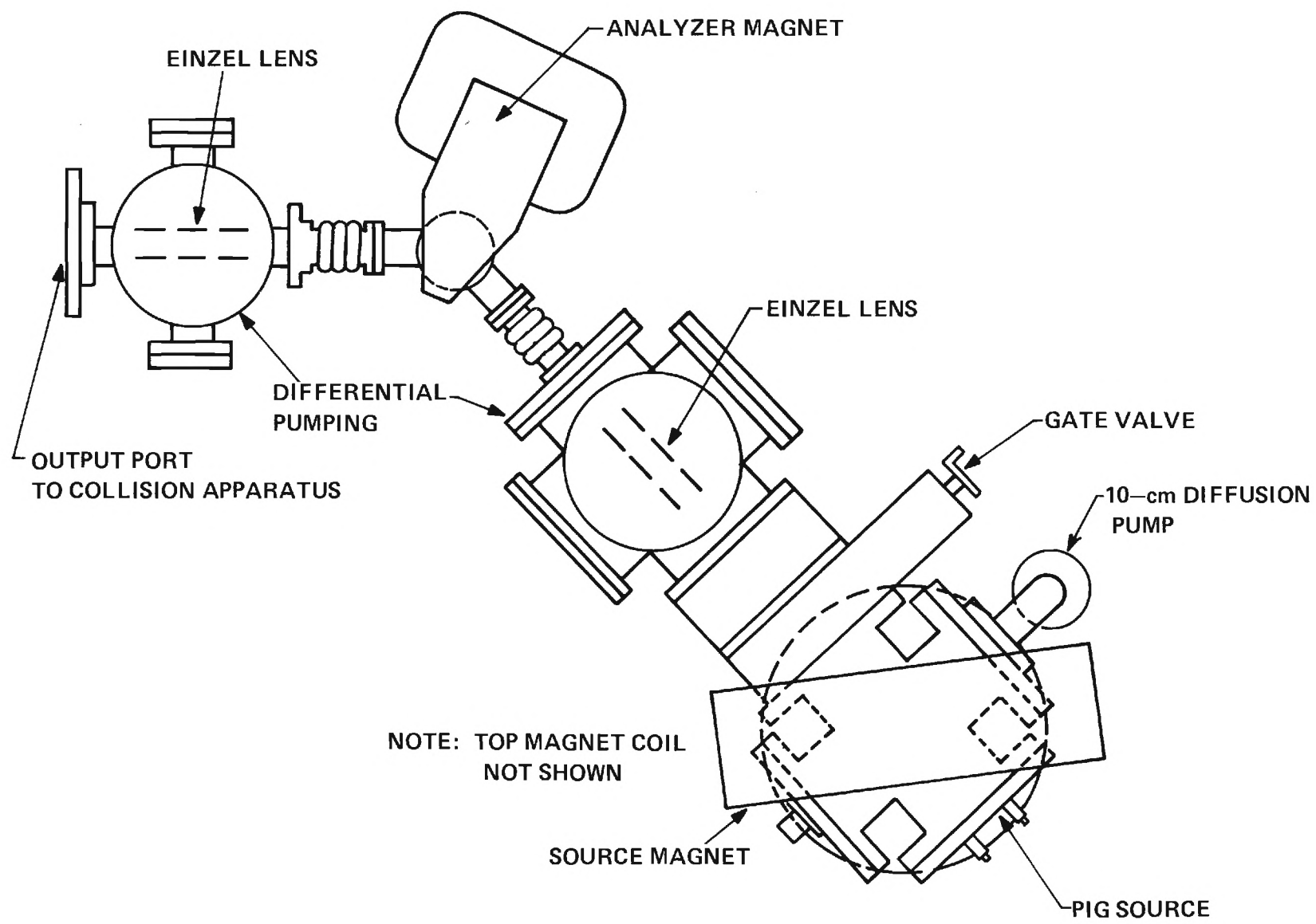


Figure A-9. Schematic Diagram of the PIG-type Ion Source and Charge State Analyzer.

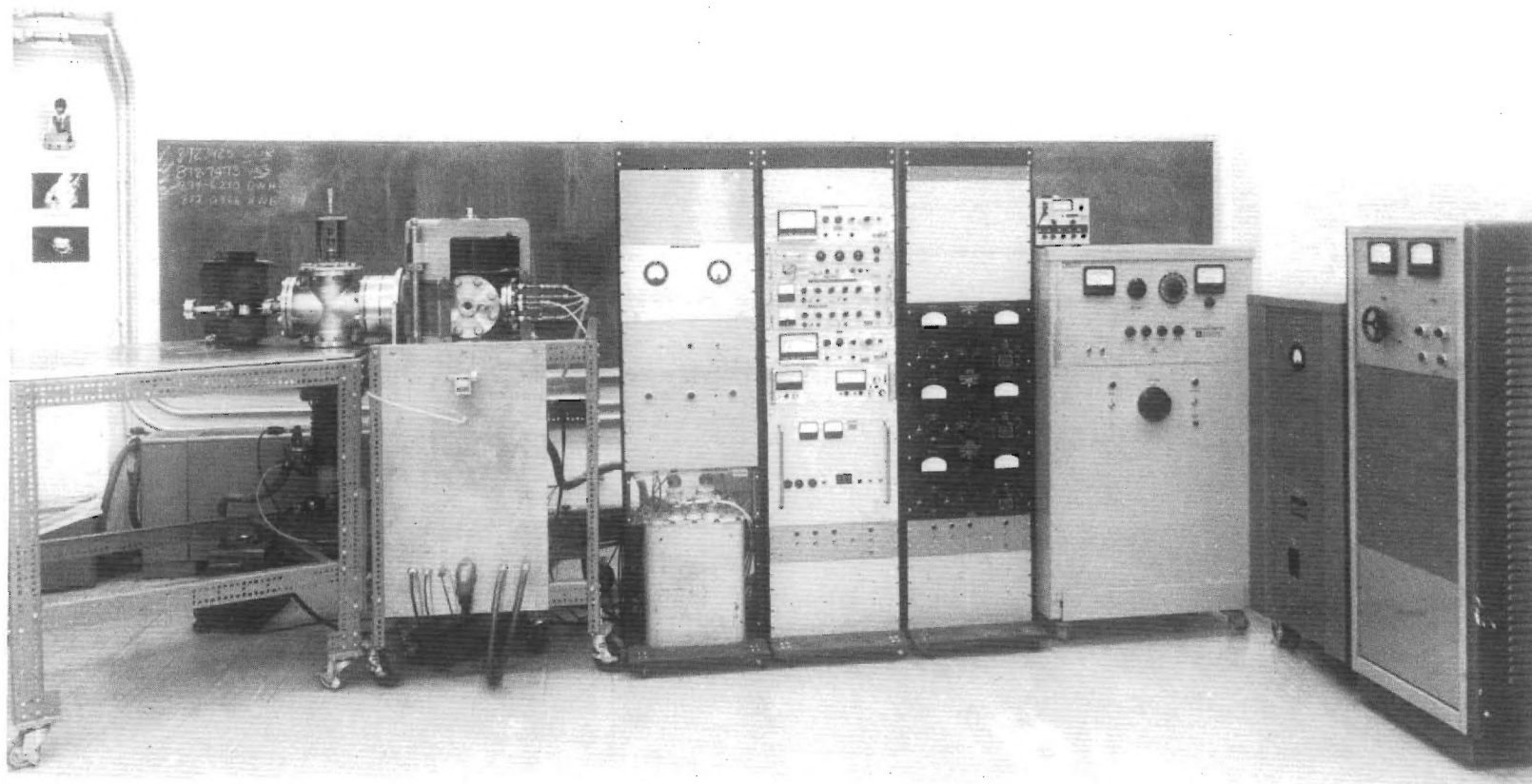


Figure A-10. Photograph of the PIG-Type Ion Source and Associated Support Equipment.

chamber. One orthogonal port is used for pumping and the third port is equipped with electrical feedthroughs and an observation window. The source vacuum chamber can be isolated from the remainder of the vacuum system by a high-vacuum gate valve attached to the ion beam exit port. This necessary feature enables periodic replacement of the PIG source without recycling the entire vacuum system.

The spacing of the six-inch diameter poles of the source magnet was originally constrained by the maximum chamber diameter. Special pole caps were machined to conform to the chamber shape. These have reduced the gap from six-inches to four-inches. This modification has increased the magnetic flux density to the desired value of about 4 kG.

Arc current is supplied by a 5 kVA plate transformer feeding a 5kV, 5A bridge rectifier. This modified power supply employs a three-ganged induction regulator to control the transformer primary current. This regulator could be used with a servo system to achieve automatic control of the arc current. However, this appears not to be required as the arc current is sufficiently stabilized by the addition of a resistor in series with the power supply. Under typical operating conditions, the PIG arc circuit draws 2-5 A at 500 V. The usual laboratory electrical service was inadequate to power the ion source, so the School of Electrical Engineering provided an additional 200A, 120/208 V three-phase service at no cost to the project.

A suitable gas is admitted through small holes near the two cathodes. Typical source operating pressures are about 10 mTorr and

may be monitored with a Baratron capacitance manometer. A significant improvement in source operation was effected by placing a stage of differential pumping immediately after the gate valve. This modification allows greater internal source pressure to exist in equilibrium with reduced source chamber pressure and has eliminated electrical breakdown problems.

Efforts during this contract period have concentrated on a thorough testing of the source. Ion yield as a function of source pressure, chamber pressure, input power and cathode design has been measured. Four different cathode configurations and two types of cathode material have been studied. Extensive spectroscopic studies in the visible and near-uv regimes have aided source optimization.

The spectroscopic techniques were used to identify those ions present within the ion source. These results were compared with the extracted ion species to provide an indication of the efficacy of the extraction process. Initial results indicated that the extractor design could be improved by changing its shape and reducing its overall length. The previously used 2 x 10 mm extraction slit was changed to a 4 mm diameter hole. This arrangement produced a circular ion beam and considerably simplified the ion beam optics. Immediately after the extractor, a set of deflection plates was inserted to neutralize the Lorentz force produced by the magnetic field. The shortened extractor allowed the deflection plates to be placed close to the source, thus reducing the deflection field requirements. A new extraction power supply was designed which could supply the current delivered by the extractor to the source plasma. An elaborate system

of electrical interlocks protected the extraction supply from various types of electrical failures.

A typical operating sequence starts with adjustment of a variable leak valve to give the desired source starting pressure. The magnetic field is then brought up to the operating value. When source gas and magnetic field are both present, the arc voltage is increased from zero until an initial breakdown occurs. This takes approximately 1.5-3 kV. The arc current is then increased until the power input to the source is about 1 kW. This power is sufficient to begin heating the tantalum cathodes. As the power is increased, it is necessary to readjust the source gas pressure in order to maintain a stable arc. An observation window into the ion source chamber was found to be essential to accomplish smooth control of the arc. When the power input reaches about 1 kW, it is noted that the arc voltage drops to about 500 V and the current increases. This is the normal operating or "negative resistance" mode. Once this mode is reached, the source power can be increased as desired, limited only by the power supply. The arc voltage remains about 500 V as the power increases. The low voltage across the source precludes electrical breakdown.

At operating powers of 2-5 kW, the cathode lifetime is less than 10 hours. The source appears to fail when the depth of an arc crater in a cathode is equal to its diameter. Several different cathode shapes have been tried in an attempt to find one which would provide superior ion yield, long life, and operate with minimum arc power. This work is continuing, and is combined with efforts to find the best configuration introducing metallic source materials.

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